Combustion theory and modeling

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1. Introduction

Theory has played an important role in combustion science for many decades. Early examples include Chapman–Jouguet detonation theory; the Burke–Schumann fast-chemistry approximation for diffusion flames (derivable in a limit process that came to be called Damköhler-number asymptotics); Frank–Kamenetskii's steady-state theory of spontaneous combustion (the origin of activation-energy asymptotics); Zel'dovich's early contributions to deflagration theory (equivalent to use of activation-energy asymptotics for achieving spatial scale separation); and the Darrieus–Landau hydrodynamic limit for deflagrations (which could be termed Peclet-number asymptotics). It is no accident that most of these examples are examples of asymptotics. Combustion problems, like those of fluid mechanics, can seldom be linearized, and so analytical strategies require mathematical tools capable of dealing with nonlinearities. Asymptotics is the only tool of universal applicability, requiring only a large parameter or coordinate for its foundation.

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Asymptotics, whether matched asymptotic expansions (such as boundary–layer theory), Poincaré–Lighthill strategies (as in perturbed orbital mechanics and sonic–boom theory) or multiple-scale techniques (justifying Krylov–Bogoliubov averaging, WKB approximations, and adiabatic invariances, for example), essentially emerged strongly after World War II (although with roots extending back to Laplace and Newton) and was vigorously developed by the fluid-mechanics community. It is difficult to pick up copies of the Journal of Fluid Mechanics from the 1950s, 1960s, and 1970s, and understand the theoretical work discussed there without at least a rough grasp of asymptotics.

In combustion, the development of asymptotics was slower and for many years was restricted to the great Russian school associated with names like Semenov, Zel'dovich, and Frank-Kamenetskii. The achievements of this school are summarized by Zel'dovich, Barenblatt, Librovich, and Makhviladze in a book [1], which in many ways is a compendium of that work. These contributions are characterized by rich physical discussion, and they challenge anyone who might feel that physical understanding and intuition are necessarily in conflict with formal mathematical strategies. It is a fact that simple mathematical models that incorporate a minimum of physics, when solved in a manner that makes transparent the physical interactions in various parts of the combustion field, and when the results are presented in a physical context, can be a source of physical insight superior to any other. It is difficult, for example, to see how the specific nature of the role of radiation in the stabilization of flame balls [2] can be correctly understood without an examination of the mathematical stability theory, while, in fact, a little thought along the lines of radiant-loss influences on flame speeds, without carefully considering Lewis-number effects, can quickly, and easily lead to an apparently plausible, but incomplete and possibly misleading picture.

If the Russian school may have had one flaw it was an apparent unwillingness, once the mathematical model was posed, to push analysis to the limits. Some hint of why this was can be found on page 369 of [1], which suggests that, given the limitations of the mathematical models (one-step chemistry, constant density, and the like), subtle, intricate details of the solution may have little physical validity. But, in fact, there is no reason to believe that the omitted physics necessarily would undo the subtle details predicted by the physics that is retained. Thus, a legitimate strategy is to push the mathematics to the limit, but be prepared to adjust the model should the solutions be at variance with the experimental record or fully detailed numerical solutions. Flame balls provide one example of rich behavior generated by a simple model consistent with the experimental record: there is a lean flammability limit [2]; one-dimensional stability only if heat losses by radiation, convection or conduction are present [2–4]; the disappearance of an interval of stable solutions as the Lewis number of the deficient reactant is increased from small values to unity [5]; three-dimensional instabilities at mixture strengths well removed from the lean limit [6]; repulsion of one flame ball by another to generate drift [7]; and stabilization by fluctuating velocity gradients of appropriate amplitudes and frequencies [8].

The theoretical papers in the Journal of Fluid Mechanics today look quite different from those of fifty years ago. These days, the applied mathematician wrestling with mechanics problems is far more likely to use scientific computation strategies than asymptotics. The same trend is now apparent in combustion (albeit this review contains counterexamples), naturally so since asymptotics has its limitations. In combustion, most asymptotic treatments are either one-dimensional or small perturbations thereof; exceptions include descriptions of the dynamics of combustion fronts for flames (such as the Michelson–Sivashinsky equation or the Kuramoto–Sivashinsky equation) or more recently for detonations, in which multidimensional combustion problems are reduced to a partial differential equation or an integrodifferential equation for a single scalar, an equation that must be solved numerically, for the most part, but a numerical task that is much simpler than the unreduced problem. It must be emphasized that the trend towards computation is not simply an abandonment of analytical strategies for computational approaches of a kind long pursued in the past. Typically, the models are still incomplete, the algorithmic investment is comparatively small, and there is an applied-mathematician's sensibility (for good and bad) that permeates the endeavor.

Recent monographs and review articles [9–26] summarize many of the main achievements in combustion theory over the past fifty years. This literature documents attainment of rather a high level of conceptual coherence. Combustion theory is, in fact, perhaps one of the most elegant areas of classical phenomenology, presenting a graphic example of the wide range of natural phenomena that can be deduced from a few fundamental principles. In the following sections, we partially complement existing reviews, focusing first on premixed flames then on nonpremixed systems, propellant combustion, turbulent combustion, ignition and detonation. We mainly look towards the future, reviewing the past only incompletely and examining its possible influence on the future.

2. Theory of gaseous deflagrations

Complexities in the analysis of deflagrations stem from the large number of elementary chemi-

cal reactions involved and from the multidimensional nature of the flow. Even in one dimension with simple model chemistry, however, the highly nonlinear nature of the conservation equations prohibits obtaining exact solutions. Activation-energy asymptotics, formally introduced in the Western literature just before 1970 [27], identifies preheat and reaction zones of different sizes, the latter small, by treating the Zel'dovich number, that is, the nondimensional activation energy $\beta \equiv E(T_a - T_u)/R^0 T_a^2$ as a large parameter, a recognition of the strong ultimate temperature dependence of the overall reaction rate. Here, E is the overall activation energy, T_a is the adiabatic flame temperature, T_u is the temperature of the fresh unburned gas, and R^0 is the universal gas constant. The resulting explicit asymptotic solution in the limit $\beta \to \infty$ provides jump conditions across the reactions zone. These account for the reactive–diffusive processes that occur on the smaller length scale within that zone. They eventually yield the burning velocity through matching. The physics represented by such a limit distinguishes combustion phenomena from other processes described by reaction–diffusion equations.

2.1. *Effects of detailed chemistry*

To account for the elementary chemical reactions that occur in deflagrations, the methods of rate-ratio asymptotics were later developed. Several steps are involved in these methods:

1. Reduce an elementary mechanism analytically to a small number of global reactions by introducing steady-state and partial-equilibrium assumptions.
2. Identify different layers within the premixed flame structure where at most two of these global reactions are active.
3. Link the layers by appropriate matching conditions to establish a complete picture of the flame structure.

For a premixed methane–air flame, the resulting structure is shown in Fig. 1. In this figure S_L denotes the laminar burning velocity, x the physical distance, and δ_L a characteristic flame thickness, D_T/S_L, where D_T is a representative thermal diffusivity. For the unstretched flame without heat loss, the final temperature T_b is the adiabatic temperature T_a.

Shown in Fig. 1 are a preheat zone having thickness of order unity in the nondimensional variable x/δ_L (as in activation-energy asymptotics), an inner layer with thickness of order δ, where the fuel is consumed and the intermediate species CO and H_2 are formed, and an oxidation layer with thickness of order ε, where those intermediates are oxidized to CO_2 and H_2O (species not shown here). The order is $1 > \varepsilon > \delta$. Embedded in these layers are additional layers, such as a radical-consumption layer [28], a hydrogen-consumption layer [29], or consumption layers of other intermediates such as C_2H_4 and CH_2O in *n*-heptane flames [30] or *i*-C_4H_8, and C_3H_4 in *iso*-octane flames [31].

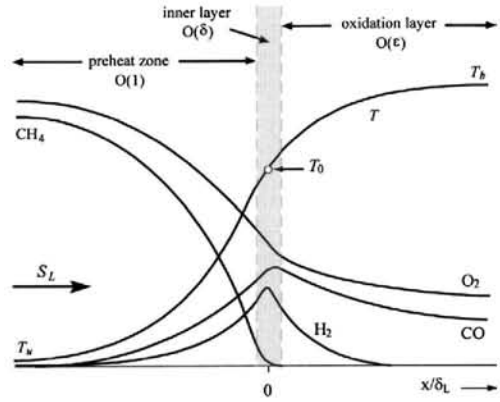

Fig. 1. Structure of a premixed methane–air flame according to rate-ratio asymptotics.

It is interesting to note that rate-ratio asymptotics of premixed flames does not provide a justification for the assumption of a large activation energy. On the contrary, this approach shows that none of the activation energies in the chemical mechanism play that role. The sensitivity of the laminar burning velocity S_L to small changes of the burned-gas temperature (which can be caused by heat loss or flame stretch, as discussed below) is, nevertheless recovered: this sensitivity reflects the relatively small temperature difference between the inner-layer temperature T_0 shown in Fig. 1 and the burned-gas temperature T_b. The inner-layer temperature may be interpreted as the crossover temperature, T_c, between chain-branching and chain-breaking reactions. It therefore is to leading order independent of temperatures of the unburned and burned gases. This finding also leads to an understanding of the lean flammability limit: when the mixture is so lean that the resulting burned-gas temperature is equal to or below the inner-layer temperature, no chain-branching, and therefore no flame propagation can occur [32].

While in lean, stoichiometric, and moderately rich premixed hydrocarbon flames, the different global reactions occurs in well-separated layers, fuel, and oxygen consumption occurs in a single reaction zone for richer methane flames [33]. Despite there being only one layer, the analysis is more complex than that for a one-step reaction occurring in a single reaction layer. Figure 2 reproduces Fig. 7 of [33] and shows comparisons of burning velocities calculated from a 61-step mechanism including C_2 species and from a three-step global mechanism with the asymptotic

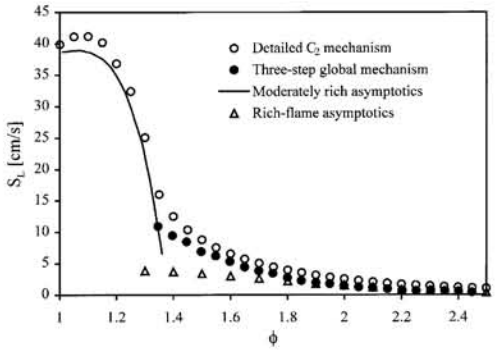

Fig. 2. Burning-velocity predictions for rich methane–air flames as a function of equivalence ratio ϕ.

results for the moderately rich [34] and rich [33] flames. Agreements are satisfactory.

These accomplishments of flame-structure analyses by asymptotic methods are impressive. Different fuels have different inner-zone structures. There is therefore much more to be done in this line of research. Advances in the future may be anticipated in clarifying premixed-flame structures of other fuels and fuel mixtures, as well as in analyzing production and consumption of trace species, some of which are of concern as pollutants, others being of possible interest in connection with synthesis of new materials. The fidelity of the results will depend on further advances needed in descriptions of transport properties [35] and on improvements in chemical-kinetic elementary-rate descriptions, uses of which were pioneered by Dixon-Lewis [36] and by Warnatz [37], for example.

2.2. *Hydrodynamic theory*

In treating multidimensional and time-dependent problems, it is helpful to integrate over the complexities of the internal structure discussed above. This leads to what has been called hydrodynamic theory. Since deflagrative combustion is normally strongly subsonic, a quasi-isobaric limit, yielding a low-Mach-number approximation, may be employed in addressing effects of multidimensionality, and flow. The flow field affects both the flame structure and its dynamics. In the hydrodynamic theory, the whole flame, associated with the region, where chemical reaction, diffusion, heat conduction, and viscous effects take place, is assumed to be thin when compared to the representative fluid-flow length scale associated, for example, with the size of the wrinkles on the flame front or with the geometrical dimensions of the vessel within which combustion takes place. The flow field is then determined by an analysis in which the incompressible hydrodynamic equations (with different densities for the burned and unburned gases) must be solved on both sides of the resulting flame sheet.

Beyond the simplest Darrieus–Landau theory, the internal structure of the flame is resolved on the smaller diffusion length scales. Asymptotic matching provides appropriate jump conditions for the pressure and velocities across the flame front as well as an equation for the flame speed, or equivalently for the flame shape. The final resulting model, still nonlinear, is a free-boundary problem supplemented by conditions that describe influences of the diffusion processes occurring within the flame zone. The model can incorporate effects of thermal expansion, differential and preferential diffusion, equivalence ratio, different reaction orders, temperature-dependent transport coefficients, transient pressure, and volumetric heat losses (radiative losses) [38–45].

The flame–speed relation derived in such models exhibits an explicit dependence on flame stretch with a coefficient termed the Markstein number. This relation has been experimentally tested in many circumstances for weakly stretched flames, such as counterflow, and spherically expanding or collapsing flames [46–56]. Measurements of the Markstein number have been performed to quantify the effects of strain and curvature. In this respect, the theory has been useful not only as a predictive tool, but also in identifying the dependence of the flame speed on stretch and of the Markstein number on the mixture composition [23,55,56], results that were found useful in correlating experimental data. Factors complicating accurate experimental tests are ambiguities in defining appropriate flame surfaces and tendencies for instabilities to develop as stretch vanishes.

Hydrodynamic models have been helpful in studies aimed at unraveling the intricate nature of flame instabilities. The complexities of the mathematical stability problem, which requires examining the response of the flame to *arbitrary* initial conditions, disturbances of *all possible wavelengths*, and a whole range of physical parameters pose a formidable task that cannot be accomplished very easily by numerical means without introducing approximations. Within the context of the hydrodynamic theory, the response of a flame to small disturbances is determined by performing an analysis of the fluid-dynamic problem in normal modes. The results yield a dispersion relation which identifies the role of the various mechanisms in stabilizing or destabilizing the flame and which provides a measure of the growth rate and of the time and length scales associated with the newly emerged structure. In this context, theory and experiments in the past fifty years have evolved hand-in-hand [57–67].

The description of the time-dependent, multidimensional flame structure that evolves beyond the instability threshold must be based on a nonlinear analysis that addresses the development of finite-amplitude disturbances. In recent years,

most of the activity in this area has been pursued in the "weakly nonlinear" regime using bifurcation theory. The description reduces, within an appropriate distinguished limit, to an explicit equation for the flame-surface evolution. This approach is advantageous for physical analysis as well as for numerical calculations. Versions that incorporate various effects such as curvature, heat loss, stretch, and vorticity production [68–72] have been derived and analyzed, revealing various characteristics of real flames. There is a need, however, to develop reliable nonlinear theories that more fully account for the interaction of the flame with the fluid flow. Since a sequence of instabilities is often a precursor to turbulence, the response of a flame to finite-amplitude distortions may provide insight into the large-scale structures observed in real flames. Analytical methods to deal with the full nonlinear problem are unlikely to become available in the near future, and advances in this area will rely on a combination of numerical computations and ad hoc modeling. An example of the latter is the extension [73] of the Michelson–Sivashinsky equation that is able to describe the small-scale structures that are generated on a spherically expanding flame surface, circumventing the limitations of the long-wavelength theory.

There are limitations to the hydrodynamic theory of premixed flames that are often encountered when trying to model flames as thin fronts. The theory requires the flame to consist of a single sheet, with no holes or folds, and it does not accommodate a discontinuity in the slope of the front—for example, the tip of a bunsen flame or the region of contact of a flame with a wall. The hydrodynamic description in these circumstances is incomplete. Instabilities can cause the flame front to develop sharp crests. When a premixed flame travels down a tube, as a consequence of hydrodynamic instability the flame is convex towards the unburned gas, with crests located at the walls, and since the flow of unburned gas is refracted by the front when crossing the flame, there is a kinematic incompatibility in the burned gas near the wall. More work is needed in this area.

Future extensions of the hydrodynamic theory are likely to include more detailed chemistry in the flame structure, of the type previously discussed. This will enable examining the influence of chemistry on the dynamics and stability of premixed flames in a more comprehensive way. For example, one may be able to identify the role of active intermediaries in flame stability. Other extensions will include modeling flames in multiphase systems, such as liquid sprays or dusty gases, where additional complexities arise because of the need to incorporate sufficient details of the dispersed-phase phenomena into the model. In these two-phase systems, depending on the particle size, particle fuel content, and volatility, various modes of burning are possible.

2.3. Different types of approaches to describing multidimensional phenomena in nonuniform flow

For understanding many aspects of flame behavior, the influence of the flame on the background flow may often be ignored, and the flow field may be regarded as prescribed. Formally, this may be achieved by neglecting thermal expansion, that is, assuming the gas density to be independent of temperature. Although in real-life systems thermal expansion is never small, the constant-density models (or models, where the density variation is accounted for but treated as a small perturbation [10]) proved to be highly successful in interpreting even rather subtle and complex effects far beyond the expected nominal range of validity of the model. At small density variations, the comparatively weaker effect of flame-generated vorticity may also be ignored, so as to enjoy the technical advantages that this assumption entails [16,26].

In problems involving flame propagation through vortical flow fields the flame interface may break up into a highly convoluted contiguous front followed by trailing islands of unburned gas. An effective way to describe the flame dynamics in these systems is by considering the level sets of an appropriate scalar field [25,74,75]. This type of approach has been employed successfully in many problems of flame-flow interaction and has even been extended to detonations.

The aforementioned are approximations for deflagrations that are derived from the first-principle conservation laws by pushing certain parameters to their limits. Yet, simplifications obtained by this strategy are sometimes still far from being analytically tractable. One may then turn to informal approximations, which cannot be obtained as distinguished limits, but which are nevertheless believed to capture the essence of the phenomenon being studied. Refs. [76–79], for example, describe essentially multidimensional processes treated by appropriately coupled one-dimensional models. This kind of ad hoc modeling requires some degree of boldness and an intuitive understanding of the physical mechanisms involved. If appropriately designed and interpreted, studied in sufficient detail and found to be robust, these models may be highly educational, providing physical insight, and unveiling unforeseen paths to more rational descriptions.

2.4. Interplay between the intrinsic instabilities and noise in wrinkled flames

The reduction of the effective dimensionality of the system and subsequent derivation of explicit equations for the flame surface were found extremely useful in discussing the evolution of pattern

formation and chaotic behaviors. There has also been significant development towards understanding the effects on flame dynamics of external noise that results, for example, from a weakly turbulent flow of relatively large scale. There are strong arguments in support of Joulin's conjecture [80] that the experimentally and numerically observed nonsteady pebbly structures in expanding and nonexpanding flames do not, in fact, represent a self-sustained phenomenon, but rather a peculiar response to an ever-present background (e.g., numerical) noise. Indeed, in the absence of noise, as has recently been shown [81,82], a single-cusp flame propagating through a channel is unconditionally stable. This obvious discrepancy with experimental and numerical observations may apparently be attributed to the impact of noise supplying small disturbances that are rapidly magnified by the hydrodynamic instability. It also suggests that other complex flows, possibly including Navier–Stokes turbulence, may not be entirely the products of initial disturbances, but might require some level of permanent noise to sustain their multiple-scale nature. The equations of flame dynamics are relatively simple, unlike those of many hydrodynamic systems, and the rational resolution of the question, either by an ingenious analytical approach or by a well controlled numerical simulation, does not seem to be an insurmountably difficult a task.

Weakly nonlinear theory will continue to be an efficient way to address problems that involve a wide range of length scales, such as flame–acoustic interactions [83,84]. For a turbulent flame, for example, the length scales involved range from the smallest Kolmogorov scale and the smallest scale associated with chemical reactions to the large wavelength of the acoustic fluctuations. Elements of the hydrodynamic theory of premixed flames were incorporated in examining effects of the acoustic field on a scale proportional to the characteristic Mach number [85,86]. These theoretical results led to the design of an experiment [87] in which different modes of amplitude amplification and attenuation were observed. A weakly nonlinear theory [88] recently provided an explanation of these modes. Challenging problems that require more judicious modeling efforts remain in addressing the fully nonlinear interaction of the acoustic field with the induced hydrodynamic field of a corrugated flame.

2.5. Possibly unexpected characteristics of deflagrations in nonuniform flows

In counterflows, periodic shear flows, and vortical flows, the strong temperature dependence of the reaction rate can cause the flame speed to be a nonmonotonic function of the fluctuation velocity, imposing an upper limit on the burning-rate enhancement. Upon reaching its maximum, with further increase in the fluctuation intensity, the burning rate drops. For mildly nonadiabatic systems, this is followed by flame disintegration and extinction. Within a purely adiabatic picture, flame extinction does not seem feasible no matter how strong the flame distortion and stretch may be [26].

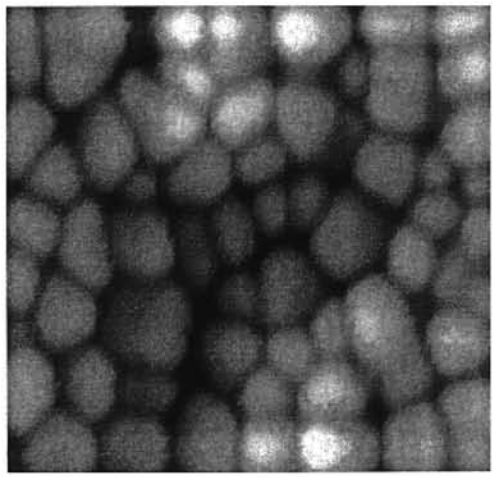

Fig. 3. Pebbly structure of a low-Lewis-number adiabatic flame in a state of chaotic self-motion, merging and splitting, from a numerical simulation of the three-dimensional constant-density reaction-diffusion model, lighter shading corresponding to more advanced parts of the reaction zone [90].

Hydrodynamic and diffusive instabilities make flames extremely rich dynamically, capable of both ordered and complex behaviors involving periodic and aperiodic pulsations, spinning patterns, chaotic self-motion (Fig. 3), inverse cascades, and a possible fractal-like growth [89]. At a sufficiently high level of heat losses, there can be self-fragmentation [90–92]. Cellular flames occurring in low-Lewis-number mixtures break up into separate cap-like fragments that sometimes close upon themselves to form flame balls (Fig. 4). The flame balls may be either stationary or self-drifting, forming, as it were, a strongly fractured cellular flame through which a considerable portion of the fuel escapes the reaction zone, and remains unconsumed [26].

3. Diffusion flames and partial premixing

As with deflagrations, during the past fifty years there have been significant advances in both activation-energy asymptotics [93] and rate-ratio asymptotics [94]. The results of these two different approaches can differ with respect to the reactant that leaks through the reaction zone and thereby causes incomplete combustion and eventually extinction. In one-step activation-energy asympt-

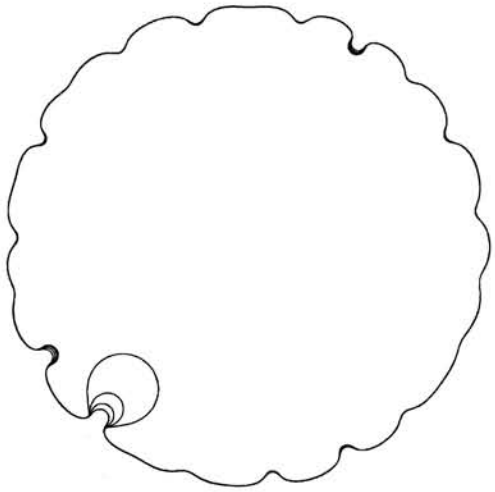

Fig. 4. Numerical simulation of a geometrically invariant model for the dynamics of near-limit cellular flames showing the inception of flame fragmentation. The flame evolution is governed by a pair of partial differential equations for the flame interface and its temperature, and the interface of an outward propagating flame is shown at four consecutive instants of time prior to blow up of the solution [92].

otics of hydrocarbon–air flames, fuel leakage is predicted, while rate-ratio asymptotics based on a four-step mechanism for methane–air flames predicts oxygen leakage, agreeing with experimental data. The latter analysis has been refined in a more recent paper [95]. Rate-ratio asymptotics was also successfully used to predict NO concentrations in methane–air diffusion flames [96] and to explain chemical flame inhibition by CF_3Br in nonpremixed methane flames [97]. Much more diffusion–flame progress by these techniques may be expected in the future.

Partial premixing leads to situations in which flames have characteristics of both premixed and nonpremixed systems. In addition, in nonuniform flows, local extinctions of diffusion flames lead to intermixing of unreacted fuel and oxidizer. In diffusion flames, because of the high sensitivity of the reaction rate to temperature, the chemical reaction will typically occur only after an ignition source triggers locally the chemical reaction. The reaction then propagates to the rest of the partially mixed flow field in the form of premixed flames, with lean and rich branches, leaving behind a nonhomogeneous mixture of combustion products with either fuel or oxygen. These reactants burn afterwards in a diffusion-controlled way in a trailing diffusion flame. In gaseous diffusion flames, the premixed flames created by the ignition source may travel all the way to the near wake of the fuel injectors, or only down to a point, which defines the liftoff height of the diffusion flame.

The premixed flames propagate, relative to the fluid, faster along the stoichiometric surface than in the surrounding regions, where the mixture is rich or lean. The structure of the flame front strongly depends on the Damköhler number, $(\delta_m/\delta_L)^2$, defined in terms of the effective thickness of the mixing layer δ_m and the thickness $\delta_L = D_T/S_L$ of the preheated zone of the stoichiometric premixed flame. The effective thickness of the mixing layer δ_m can be defined in terms of the rate of scalar dissipation of the mixture fraction Z, evaluated at $Z = Z_S$ the stoichiometric surface, namely $\chi_S = D_T(\nabla Z)_S^2$, as $\delta_m^2 = D_T/\chi_S$, or for small Z_S more precisely $\delta_m^2 = Z_S^2 D_T/\chi_S$.

When $(\delta_m/\delta_L)^2$ is large compared with unity, the flame front has a triple-flame structure, there being two premixed flames that meet at a diffusion flame. For large values of the effective overall activation energy β of the reaction, the characteristic scale of the reaction front is δ_m/β, which may be compared with δ_L to define an effective Damköhler number $Da = (\delta_m/\beta\delta_L)^2$. The flame speed U_F measured against S_L is a function of Da and mainly depends on two parameters, s, the mass of the airstream required to burn a unit mass of the fuel stream and r, the ratio of the stoichiometric and initial air temperatures, which characterizes the effects of the exothermicity of the reaction. For values of Da moderately large compared with unity, U_F/S_L becomes independent of Da and is a function of these two parameters, as well as of β, albeit weakly if β is large.

When $(\delta_m/\delta_L)^2$ becomes of order unity, the two premixed flames merge with the diffusion flame, forming a single flame with an edge. Illustrative of this topic being a very active area of current investigation is the lack of standardized terminology. Here, we call the structures for large values of $(\delta_m/\delta_L)^2$ triple flames and those when this quantity is of order unity or smaller edge flames. Some prefer instead to introduce the compound noun edge-flame, or sometimes flame-edges, often using the former for the entire range of $(\delta_m/\delta_L)^2$, so that the triple flame (which some prefer to call a tribrachial flame) becomes a subcategory. Confusion of this kind is typical of a hot area of investigation.

3.1. Triple flames

A representative triple-flame configuration is that downstream from the edge of a splitter plate, as illustrated in Fig. 5. In this case, the flame is stabilized at a distance from the tip by heat loss to the plate. Although most triple-flame calculations address symmetric problems ($s = 1$) in a constant-density approximation ($r = 1$), the thermal expansion associated with the exothermicity of the reaction introduces displacement velocities of order S_L by the premixed flames in the upstream region and of order D_T/δ_m by the diffusion flame downstream. Those associated with the premixed

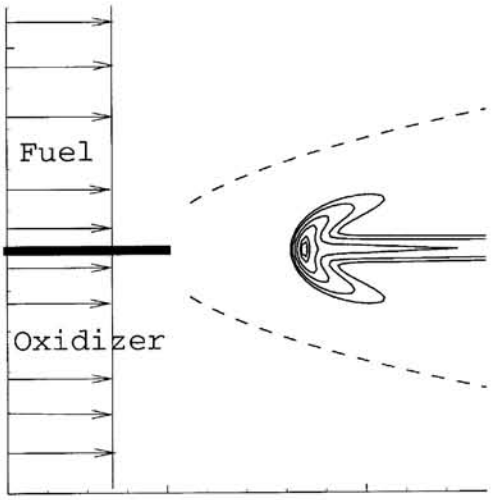

Fig. 5. A representative triple flame, shown by contours of constant values of the chemical reaction rate, from M. Matalon.

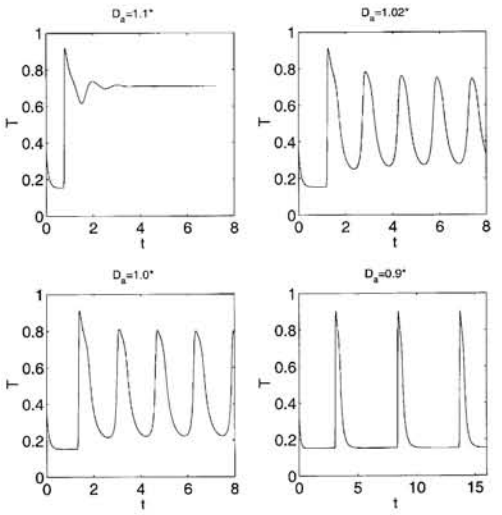

Fig. 6. The nondimensional temperature history at a point in the combustion field for an anchored edge flame having a fuel Lewis number of 1.8 and an oxidizer Lewis number of 1.0 for various Damköhler numbers [103].

flames result, for large values of Da, in a ratio U_F/S_L of the order of $\sqrt{r}$ when r is large. Triple flames have been addressed both analytically (e.g., [98]) and numerically (e.g., [99]), in configurations ranging from counterflows to mixing layers to jets; nevertheless, there still is more to be learned about these flames.

3.2. Edge flames

Edge flames, encountered at smaller Da, where $(\delta_m/\delta_L)^2$ is of order unity or smaller, constitute another area in which scientific computation of the simplified kind mentioned in the introduction has played an important role. Although the underlying conservation equations are complicated in that at least two dimensions are involved, there is a one-dimensional model [100] that retains much of the physics, and affords analytical simplifications that have been exploited [78,101]. Depending on the value of $(\delta_m/\delta_L)^2$, flame edges may advance or retreat. The former is often called an ignition front and the latter a failure wave. One-dimensional models provide insight into such behavior and enable reaction-diffusion types of equations to be derived for time-dependent motions of edges that may not be straight.

Stability analyses for edge propagation are relevant [78]. Edge flames can display pulsations, even when the trailing structure is stable [102,103]. They can also develop cellular instabilities [104,105]. For edge flames stabilized by a splitter plate, at high flow rates oscillations occur when at least one of the two Lewis numbers is sufficiently large, and oscillations can be enhanced by heat loss [106]. Figure 6 illustrates how the oscillations develop and become more pronounced as the Damköhler number decreases. Cellular behavior is illustrated in Fig. 7.

In addition to occurring near boundaries, diffusion-flame edges can be encountered away from boundaries. For example, when a flame–vortex interaction stretches the diffusion flame with a nondimensional stretch rate $\chi_S\delta_L/S_L = (\delta_L/\delta_m)^2$ above a critical value of order unity, local flame extinction occurs. A hole with edges then appears in the diffusion flame. The hole grows rapidly in size if the stretch rate is maintained, because close to the extinction value of Da, of order $1/\beta^2$, the

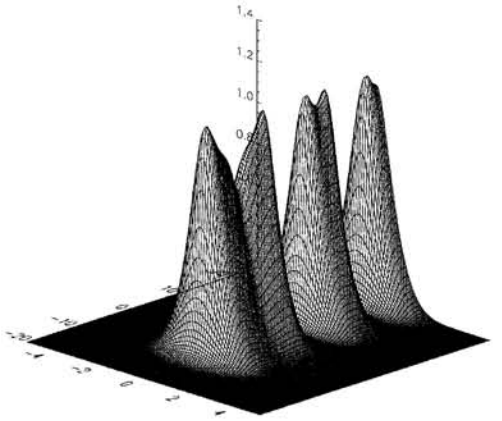

Fig. 7. The temperature topography of a diluted hydrogen–air edge flame moving to the left, in which the post-edge structure splits to leave a train of stationary flame strings [107].

edge front velocity U_F/S_L is negative and, more importantly, the flame-edge displacement is assisted soon by the flow velocity components associated with the stretch. The hole collapse will occur only when the stretch rate is decreased again [108].

Diffusion-flame edges are also encountered in nonpremixed flames attached in the near wake of the fuel injector, when the fuel (or air) boundary–layer velocity gradient, measured with S_L/δ_L, is smaller than a critical value of order unity. Since upstream diffusion and heat conduction to the injector wall play an essential role in anchoring the diffusion flame, analysis predicting liftoff requires numerical solution of the compete reactive Navier–Stokes equations [109]. The structure of the edge is strongly influenced by the local velocity gradient.

The spreading of a diffusion flame over a solid fuel burning in also air can be controlled by edge-flame propagation [110]. Edge flames can also arise in a premixed context in a flow of varying strain rate [107,111–113]. Since there thus are many configurations in which edge flames are relevant, extensive studies of edge flames are likely to appear in the future.

4. Combustion of solid propellants

So far we have discussed models that look very much like those used in analytical strategies, but in more complex problems, such as those encountered in studies of the combustion of solid propellants, the key creative ingredient is the formulation of the model. Once this is correctly accomplished, all that remains in principle are calculations and interpretations of the results. For much numerical work, however, the challenge lies in the numerics, and the model is nothing more than a conventional expression of presumed known physics. In these situations, the theoretician's role is driven by the needs of computation. For heterogeneous solid propellants, this is a challenging numerical problem that requires the solution of an unsteady three-dimensional combustion field coupled, by an unsteadily regressing nonplanar surface, to an unsteady thermal field in the solid. A number of difficult theoretical challenges have to be met with before a suitable code can be developed.

A representative propellant consisting of ammonium perchlorate (AP) particles embedded in the fuel binder hydroxyl-terminated polybutadiene (HTPB) has particle sizes ranging from hundreds of microns to one micron, constituting roughly 80% of the volume of the propellant. If the particles are represented by spheres, a packing algorithm is needed with which a model morphology can be constructed with the essential characteristics of real propellants. An effective way of doing this is to introduce a particle-dynamics strategy [114].

In a typical propellant pack, the smallest AP particles cannot be numerically resolved and yet can occupy a significant fraction of the volume. To account for these, homogenization formulas are needed, one for obtaining the effective thermal conductivity of an AP–HTPB blend, another for the effective pyrolysis law for a blend. Homogenization formulas for heat conduction can be derived by hypothesizing that if an additional AP particle is added to a pack, the change in conductivity is the same whether the addition is incorporated as a blend or as additional discrete particles [115]. The resulting homogenization equation is

$$1 = (1-t)^3 \frac{1-x^3}{F-x} F,$$

where $F = F(x,t)$ is the effective conductivity normalized with the conductivity of the binder, λ_B, t is the packing (volume) fraction of AP particles, and $x = \lambda_{AP}/\lambda_B$, where λ_{AP} is the conductivity of the AP.

The propellant surface consists of either AP or HTPB, each of which is converted to gas at different rates. A common way of describing the conversion is through a pyrolysis law such as the Arrhenius law with activation energy E_i and prefactor A_i, which can be written as $r_{b,i} = A_i \exp(-E_i/R^0 T_s)$, where $r_{b,i}$ is the surface regression rate and T_s is the surface temperature. An appropriate homogenization formula for the regression rate of the blend here is [115] $r_b = r_{b,AP}^t r_{b,B}^{1-t}$, also an Arrhenius law.

Another role for theory emerges after the full three-dimensional code has been assembled. Although the latter can be used to predict burning rates for different propellant morphologies, it cannot be coupled with a code that describes the rocket chamber flow, because of the large difference in length scales. Instead, the description may be averaged to generate a one-dimensional description. This involves identifying and modeling averaged terms that contribute to the solid-phase heat conduction and constructing a look-up table from the three-dimensional calculations, so that the heat flux from the combustion field to the solid can be correctly described. In this example, what at first sight appears to be a massive numerical problem, to be addressed exclusively by highly skilled code writers, cannot be solved without significant input from theoreticians, modelers with a mathematical bent.

There have been many other important contributions to the theory of heterogeneous propellant combustion over the past fifty years, some of which are described in a monograph [116]. This is a difficult area of research to which theoreticians may be expected to make important contributions in the future. The same may be said for homogeneous propellant combustion, which has

also experienced recent advances in theory, including unsteady phenomena [117–119]. One aspect of homogeneous propellants is that, over time, they degrade, and become porous. Progress has been recently made in the theory of deflagration of porous propellants [120]. Filtration combustion is a related topic in which excellent progress in detailed theory has been recently made [121]. Other related topics to which considerable attention has been paid include coal combustion and devolatilization [122,123], and material flammability and combustion [124,125]. The combustion of solid materials, involving multiphase phenomena, is a very rich field of study in which extensive future theoretical advances should occur.

5. Turbulent combustion

Results from laminar flame studies have inspired the modeling of turbulent combustion ever since its beginning by Damköhler [126] in 1940. In 1982, asymptotic multiple-scale analyses [39–41] predicted that the laminar burning velocity depends on flame stretch if the Lewis number is different from unity. Consequently, a stretch factor was introduced in [127] to modify the source term in the equation for the mean progress variable. In [128], experiments were re-analyzed, demonstrating that Lewis numbers different from unity modify the turbulent burning velocity. The basis for this reasoning is the flamelet concept that views a turbulent flame as an ensemble of stretched laminar flamelets, defined as asymptotically thin layers [13] embedded within the turbulent flow field. The multiple-scale approach was emphasized in [25] for both premixed and nonpremixed turbulent combustion. Other aspects are discussed in multi-author reviews [129,130].

For premixed combustion, asymptotic flame-structure results helped one to define different regimes, including the corrugated-flamelets regime, where the entire flame structure is thin compared to the Kolmogorov scale, and a regime called the "thin-reaction-zone regime," where small eddies of the size of the Kolmogorov scale are able to enter into the preheat zone shown in Fig. 1, but not into the inner layer, so that the reaction zone remains thin compared to all scales of the flow.

Various modeling issues of premixed turbulent combustion were clarified by using the level-set approach based on the G-equation [131,132]. Scaling arguments [133] showed that in the corrugated-flamelets regime both strain and curvature effects are of higher order because they are active at smaller scales than the Gibson scale, the cutoff scale determined by the burning velocity itself. Curvature effects, on the other hand, become dominant in the thin-reaction-zone regime, where the cutoff scale is determined by the laminar diffusivity [134]. A previous analysis [135] of the response of one-dimensional premixed flames to time-dependent stretch and curvature had shown that Lewis-number effects disappear at high frequency such that the Markstein diffusivity becomes equal to the mass diffusivity. This result was used in [134] to postulate a common G-equation for both corrugated-flamelet and thin-reaction-zone regimes. As a result, a more fundamentally based expression for the turbulent burning velocity could be derived. It attributes the bending of the turbulent burning velocity, when plotted as a function of the turbulence intensity, to diffusive effects associated with small-scale flame curvature.

There are many unresolved problems in premixed turbulent combustion that could be addressed on the basis of direct numerical simulations (DNS) of the G-equation in a turbulent flow field. The advantage of such an approach is that, given the heat release, the influence of chemistry is parameterized by fixed values of the laminar burning velocity and its response to strain, curvature, and potential heat-loss effects. The unresolved question of flame-generated turbulence, considered in [136], should be studied in more detail to propose rules for modeling.

From the practical point of view, large-eddy simulations (LES) based on the G-equation present further advantages over the classical Reynolds-Averaged Navier–Stokes (RANS) approach [137]. By taking averages over smaller regions in the vicinity of the flame front, the inherent front instabilities and the interaction caused by gas-expansion effects between different parts of the front can be captured. More work in this area is certainly encouraged.

As far as nonpremixed combustion is concerned, alternative formulations of mixture-fraction-based models have stimulated the discussion over the past fifteen years. While the flamelet concept [138] may be justified by two-scale asymptotic analysis [25], the conditional moment closure (CMC) formulation [139] relies on turbulent closure procedures more challenging than those of RANS. It is interesting to note that the leading-order unsteady flamelet equations and the equations for the first conditional moments are identical, if the same order-of-magnitude assumptions for the different terms are made. In practice, higher-order formulations of the flamelet equations may be as impractical as higher-order moments of CMC. Both approaches therefore shed a different but reassuring light on mixture-fraction-based models.

The weakness of these models, however, lies in the modeling of their most important parameter, the one that couples the flamelet-CMC equations to the turbulent flow field, the conditional scalar dissipation rate. Fluctuations of this quantity

have large effects on pollutant predictions in turbulent-jet diffusion flames, as recently shown by LES simulations [140]. While extinction of burning diffusion flame structures and autoignition of mixing layers can be easily predicted in terms of threshold values for the scalar dissipation rate, unsteady local extinction and reignition events in nonpremixed turbulent combustion are not well predicted or even understood. Once a diffusion flame sheet is broken by local extinction, unsteady premixed-flame propagation of the edge-flame type may close the hole if the scalar dissipation rate attains a sufficiently low value. DNS studies will certainly help one to sharpen the understanding of these phenomena, but in the end physical modeling will be needed [141]. Such modeling needs to recognize the close link between diffusion and reaction, similar to the way it appears in the expression for the laminar burning velocity, and conventional pdf-transport-equation models, which separately model chemistry and molecular mixing, the latter being based on the integral turbulent time scale, are unable to satisfy this requirement.

6. Ignition theory

Ignition of homogeneous mixtures using a one-step reaction with a large activation energy is a classical field that goes back to the Russian literature in the 1920s. More recently, rate-ratio asymptotics was applied to hydrogen–oxygen [142,143], methane–oxygen [144], and *n*-heptane–air [145] mixtures. Acetylene [146] and ethylene [147] have also been treated in this manner, although somewhat less rigorously because of the greater complexity. Different regimes of ignition were analytically identified in these studies. Furthermore, crossover temperatures, T_c, where branching and recombination rates are equal, and the rate-determining global and elementary reactions for each regime, as well as effective overall activation energies, often a weighted product of activation energies of some rate-determining elementary reactions, were determined. This kind of analysis complements numerical studies of autoignition in homogeneous mixtures, which are easier to perform and, with respect to the many details of large reaction mechanisms, more complete. Its strength, however, lies in the analytical predictions that it provides, independent of particular initial conditions, and associated simplified physical insights into ignition processes. More analysis for different fuels along this line is to be encouraged.

An important next step in this area is the analysis of ignition in one-dimensional nonuniform mixtures. In the framework of one-step activation-energy asymptotics, ignition of mixing layers with unsteady changes of strain and pressure has been analyzed for small density changes [148] and for density changes of order unity [149]. Global rate parameters were determined by comparison with experimental data from ignition in a counterflow configuration. It would be worthwhile to extend these analytical studies to multistep rate-ratio asymptotics. Steps in this direction have been taken for hydrogen–oxygen systems [150,151], and more attention is required for other fuels.

7. Theory of gaseous detonations

The past fifty years has been a period of remarkable advancement in theory of gaseous detonations. The concept of the steady, planar ZND wave structure was known before 1950, but its instability was not known. Earlier reviews [152–155] document the discovery of the prevalence of cellular and galloping detonations that result from this instability and the initial steps towards developing theoretical understanding of them. Pioneering stability analyses [156–160] laid the groundwork for later, more detailed investigations of the characteristics of the instabilities.

The most relevant temperature for studies of detonation stability is the Neumann temperature T_n that exists immediately behind the leading shock. In many situations, detonability limits correspond to equating T_n to a crossover temperature T_c, detonation not occurring if $T_n < T_c$. One-step Arrhenius reaction-rate approximations, which have been of some use in studying detonation instability, do not easily lend themselves to identification of an effective T_c. Detailed detonation chemistry with $T_n > T_c$, in general can be described as an induction stage of duration τ_i during which $T \approx T_n$, followed by a rapid runaway during which appreciable heat release begins to occur, which, in turn, is followed by a heat-release stage, the duration of which typically is comparable with τ_i. The duration of the heat-release stage depends only weakly on temperature, while τ_i is strongly dependent on T_n, having E/R^0T_n typically of order ten in an Arrhenius approximation. This character can be captured by extending two-step (branching, recombination) deflagration descriptions to three steps through addition of an initiation step (unimportant in deflagrations, but essential for the detonation induction period)—an approximation useful in addressing detonation [161,162].

7.1. Pulsating detonations

Although theoretical studies of one-dimensional time-dependent pulsating detonations (focused on interactions between shocks and reaction regions described by reactive Euler equations) are relevant to galloping detonations, their

main contribution is as a preliminary step towards understanding cellular structures and other unsteady phenomena in gaseous detonations. Besides recent numerical investigations [163–166], there have been nonlinear analytical studies of overdriven waves making use of the limits of large propagation Mach number, M_u, and specific-heat ratio, γ, near unity (approaching a Newtonian limit in which the Mach number at the Neumann state is small) [167,168]. The pulsating instability is not purely thermoacoustic but rather convective-acoustic, a perturbation at the shock being convected downstream (as an "entropy wave"), where the reaction causes a pressure disturbance propagated acoustically back upstream. The analytical work resulted in a nonlinear integral equation for the time evolution of T_n, the integral coming from the time delay of the downstream convective transport of a fluid element through the induction zone.

Linearization for small departures of T_n from its steady-state value, in a distinguished limit of strong sensitivity of the heat-release rate to T_n, rendered the integral equation linear and resulted in identification of a bifurcation parameter, having the property that the steady solution becomes unstable when this parameter exceeds a critical value [167]. A different expression for this bifurcation parameter was derived on the basis of a further analytical study that treated detonations near Chapman–Jouguet conditions in a limit of heat release small compared with the thermal enthalpy at the Neumann state [169]; the main time delay becomes the upstream acoustic-wave propagation time as Chapman–Jouguet conditions are approached. A resulting suggested general expression of the condition for instability to occur is

$$\frac{\gamma-1}{2}\frac{E}{R^0 T_n}\sqrt{\frac{Q}{c_p T_n}} > cM_n\sqrt{f},$$

where Q denotes the heat release per unit mass of mixture, c_p the specific heat at constant pressure, M_n the Mach number at the Neumann state, and f the overdrive factor, defined as the ratio of M_u^2 to the square of the Mach number of propagation of the Chapman–Jouguet detonation. Here, E is an overall activation energy measuring the sensitivity of the heat-release profile to T_n, and c denotes a constant, typically of order unity, the value of which depends on the shape of the heat-release profile. The result indicates that large γ, activation energy, and heat release favor instability, while large overdrive can stabilize the detonation to pulsation.

The shape of the heat-release profile is relevant in that, if the heat is released too quickly after the induction period, then c becomes very small, and the detonation is always subjected to pulsating instability. In the limit of instantaneous heat release, the so-called square-wave model, the integral equation becomes an equation with a negative time delay, equivalent to a difference-differential equation of the advance (as opposed to delay) type, resulting in an infinite spectrum of discrete unstable modes having growth rates increasing unboundedly with increasing frequency, an entirely unrealistic result. For realistic heat-release profiles, after bifurcation the nonlinear integral equation predicts pulsations of increasing amplitude and irregularity with decreasing overdrive, as illustrated in Fig. 8, which was actually obtained from numerical solution of the differential equations in a one-step, Arrhenius approximation [167]. This figure shows an approach to rather chaotic behavior at $f = 1.2$ and dynamic extinction at $f = 1.1$, not associated with a crossover detonability limit. Further research on this topic is warranted, for example in testing the conjecture posed in the previous equation and in exploring possible relationships to experiments on galloping detonations.

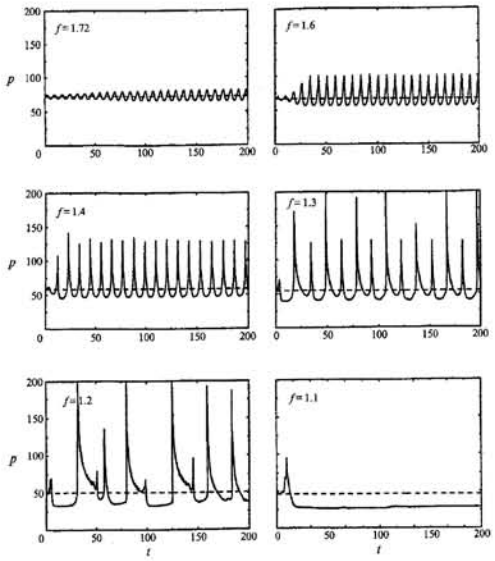

Fig. 8. Evolution of the pressure at the Neumann spike normalized by the initial pressure p_u obtained by direct numerical simulations with different degrees of overdrive and a fixed value of the activation energy in the Arrhenius law [167]; the horizontal scale is the time divided by the half-reaction time.

7.2. Cellular detonations

Significant progress has also been recently made in understanding the multidimensional instabilities that lead to cellular detonations. Although the temperature-sensitive pulsations described above play a role, there is an additional source of multidimensional instability, even for totally temperature-insensitive chemistry, pro-

vided only that there is finite-rate heat release, as, of course, there always must be in detonations. This additional effect involves transverse propagation of acoustic waves in the compressed reacting gas but is caused by nearly isobaric density changes—a hydrodynamic phenomenon associated with perturbations in the heat-release distribution caused by streamline deflections across the leading shock [170]. Compressibility effects can counterbalance this instability for sufficiently small heat release [171]. Numerical studies have helped one to clarify the linear stability spectrum [172,173] and nonlinear behaviors [174–177].

Since the Mach number at the Neumann state is small, and transverse acoustic-wave propagation behind the leading shock is relevant, transverse cell sizes will be large compared with the induction-zone thickness [170]. A weakly nonlinear analysis in the neighborhood of the stability threshold [178] remarkably reproduces patterns resembling cell structures experimentally observed, as may be seen from Fig. 9. This analysis, which makes use of limiting approximations analogous to those indicated above, results in a quadratically nonlinear partial–differential integrodifferential equation for evolution of the leading shock. It is an expansion about the stability limit obtained [171] when $Q/(c_pT_n)$ is of order $(\gamma - 1)$, so that $Q/[(\gamma - 1)c_pT_n]$ is of order unity. The result is different from that obtained from a stability analysis [179] based on a limit in which the last of these parameters is small. Further information on these developments is available in a recent review [180]. These recent studies demonstrate that a great deal of understanding can be gleaned from careful analyses of different asymptotic limits. There is more worthwhile research to be done in the future along these lines, addressing different limits, and extending results to strongly nonlinear triple-wave interaction patterns experimentally found in cellular detonations. Better understanding of conditions for occurrence of regular and irregular cells would be one important aim of future work. More study of transitions to spinning detonations (the general characteristics of which have been explained rather well in work during the past fifty years) could also be of interest in the future.

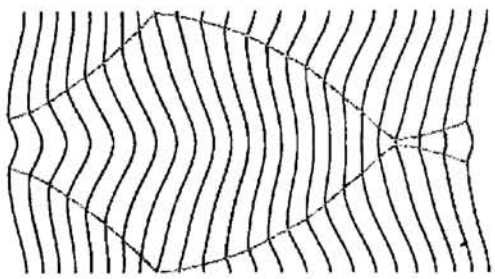

Fig. 9. Representative shock-front evolution calculated from a weakly nonlinear theory, with cusp trajectory indicated [178].

7.3. Direct initiation

A detonation may be directly initiated by an energy source of a sufficiently large intensity. Let E_0 denote the quantity of energy that is deposited. When the deposition time is small compared to the acoustic time scale τ_a (for the sound to cross the region of deposition, $\tau_a \approx r_0/a$), the flow takes the self-similar form of a Taylor–Sedov inert blast wave initiated by an ideal point source. This approximation is accurate at intermediate distances, for a radius r larger than the size of the region where the energy is initially deposited, but smaller than a typical radius for which the heat release is of the same order of magnitude as the energy initially deposited, $r_0 \ll r \ll R(E_0)$; in spherical geometry, $R(E_0) \approx (E_0/\rho_0 Q)^{1/3}$, Q denoting the heat release per unit mass and ρ_0 the initial density. The strength of the blast wave is an increasing function of E_0. At later times, when the radius of the leading shock increases and approaches values of the order of $R(E_0)$, the heat release can no longer be neglected, the blast wave triggers first a strongly overdriven detonation, and different subsequent regimes are identified from experiments [154,155]. Below a critical energy, $E_0 < E_c$, the strongly overdriven detonation decays rapidly, and the reaction front eventually separates from the leading shock, finally resulting in a premixed flame that trails behind the inert shock, and no detonation is initiated in the cold mixture. For $E_0 > E_c$, the overdriven detonation relaxes to an expanding Chapman–Jouguet detonation. The onset of the Chapman–Jouguet wave occurs at a radius of order $R(E_0)$. No Chapman–Jouguet detonation can be observed with a front radius smaller than a critical radius defined as $R_c \equiv R(E_c)$.

Well-documented experiments [152,155,181] show that, contrary to early predictions [182], R_c is not of the same order of magnitude as the largest intrinsic length scale in the problem, the total reaction length L_0 of the planar Chapman–Jouguet wave, but is instead larger than that length by two or three orders of magnitude, $R_c/L_0 \approx 10^2$–10^3. Motivated by apparent similarities in length scales, researchers proposed an empirical correlation relating R_c to the cell size [154,155]. A later theoretical analysis [183] suggested a different viewpoint. Analyses of structures of quasi-steady curved detonations are relevant here [184].

In a phase plane of the ratio of the detonation velocity D to that of a planar Chapman–Jouguet wave D_0, as a function of R/L_0, there is a saddle-like behavior, with two limit solutions (both having $D/D_0 < 1$) existing for R/L_0 above a critical value, R_c/L_0, such that fully subsonic-flow

solutions exist for values of D/D_0 above the larger and below the smaller of the limiting values, but no solutions exist for D/D_0 between the limiting values [183,184]. For $R/L_0 < R_c/L_0$, fully subsonic solutions exist for all D/D_0, and rarefaction waves from the direct-initiation position can overtake the detonation and weaken it, finally causing it to fail. For $R/L_c > R_c/L_0$, the larger of the two limiting solutions is an attractor, the detonation eventually approaching this solution at long time. A first approximation to E_c can therefore be obtained from the quasisteady critical value R_c [183]. Analysis with a square-wave model for large activation energy results for spherical geometry in $R_c/L_0 = (E/R^0T_n)(24e\gamma^2)/(\gamma^2 - 1)$, with the corresponding detonation-velocity deficit $D_0 - D_c = (D_0/2)(R^0T_0/E)$. Typical values of the right-hand side of this equation are of the order of 10^2, in rough agreement with empirical correlations. These values, which arise from the gas dynamics, geometry, and quasi-one-dimensional wave structure, do not involve cellular detonation structure at all. Time-dependent phenomena that are not quasisteady introduce some modifications, particularly in planar configurations where dynamic quenching has been computationally observed. Numerical studies with more complex chemistry [185,186] such as a three-step kinetic model [187,188] support these conclusions.

These recent advances in our understanding of direct initiation are notable. There have also been advances in understanding of initiation by nonuniform preconditioning (soft initiation) [22,26], in which initial gradients of the induction time result in spatial nonuniformity of pressure growth that can lead to the formation of compression waves capable, under favorable conditions, of triggering detonation. Further progress of knowledge in these areas is to be anticipated in the future.

7.4. Transition from deflagration to detonation

Gradients of the induction time also exist in the process of transition from deflagration to detonation. While suitable gradients can produce detonation quickly [26,189–192], they can also quench detonations under certain conditions [192]. This emphasizes the complexity of the transition process. Much remains to be learned about transition. Turbulent combustion, experimentally observed and often considered to play a dominant role in transition, cannot tell the whole story because turbulent burning velocities are still a factor of 10 or more too small to generate strong enough pressure waves. Studies of well-conceived model problems can do a great deal to improve comprehension of transition.

Hydraulic-resistance models provide one attractive means for investigating phenomena associated with transition [26]. In such models, it is found that there is a multiplicity of detonation regimes [26,193]. The hydraulic resistance gives rise to a powerful agency (diffusion of pressure) capable of driving the combustion wave both at fast subsonic as well as supersonic velocities. The latter mode may be relevant to the so-called choking regime occasionally observed in obstructed channels and smooth-walled capillaries [194–196].

Deflagration-to-detonation transition in hydraulically resisted flows [26,197] can be considered from the viewpoint that the hydraulic resistance (friction) causes a gradual precompression and, hence, preheating of the fresh mixture adjacent to the advancing deflagration. After some induction period, this development leads to a thermal explosion, triggering an abrupt transition from deflagrative to detonative propagation. The detonation first develops in the boundary layer, where the impact of hydraulic resistance is stronger and thereafter spreads over the channel interior. The second stage of the transition, however, does not proceed gradually, but rather it develops through a localized autoignition within the interior of the accelerating tulip flame that replaces the incipient dome-shaped flame.

Despite all the recent advances in understanding and description, there are quite a few aspects of the transition in need of further research. These include the origin of the elongated tulip flame accompanying the predetonational acceleration, the role of heat losses and wall roughness, identification of the major factors controlling the predetonational time and distance, description of different modes of transition (transition within the flame brush, on the leading shock, on the contact discontinuity, etc.), and identification of agencies facilitating and inhibiting transition.

Deflagration-to-detonation transition in unconfined systems is more problematic. There are reports claiming that in highly sensitive oxygen-based mixtures the transition may be triggered by outwardly propagating 'free-space' flames [198–200]. In this description, the transition is commonly attributed to the flame acceleration induced by the Darrieus–Landau instability. Yet, the acceleration resulting from wrinkling seems to be rather a weak effect whose ability to cause the transition is not at all obvious. Moreover, there is an opinion that in truly unconfined systems transition is actually unfeasible [196]. Much more research, both experimental and theoretical, is needed on this topic.

Also warranting further study is the relationship of deflagration-to-detonation transition to knock in spark-ignition engines. The unburned mixture is compressed by the engine piston as well as by the burned-gas expansion. The resulting increase in the unburned (end-gas) temperature often leads to its spontaneous ignition and pressure pulses. Detonation is to be avoided since

the emerging pressure peaks can be ruinous to the engine. The end-gas autoignition seldom occurs uniformly throughout the charge but rather arises at localized exothermic centers (hot spots) preferentially emerging near the wall. The reason for this localization is not, as yet, well understood, and the topic is still an area of controversy and numerous conjectures, involving various aerothermochemical arguments [26,201–204].

8. The future

Whither combustion theory in the next fifty years? The impossibility of formulating a rational response for such an extended period goes without saying. Fields of scientific inquiry are born, grow, and prosper, then eventually begin to decay as the body of knowledge that they can usefully generate begins to saturate. So it shall be with combustion theory, as it has been in the past with a number of related but more specific topics, such as hypersonics, water-wave theory, linear elasticity, and special-function theory of mathematical physics. The questions to be addressed concern time scales. Why, after more than sixty years of activity, does combustion theory remain so vigorous? When will the intensity of investigation begin to subside?

An underlying cause for the longevity of the field ultimately stems from the character alluded to in the final paragraph of the introduction—a plethora of exciting phenomena emerge from the basic principles of the subject. Another contributing factor has been the technological needs of energy, propulsion, weapons, pollution mitigation, and safety hazards, which have helped to encourage support for research. For these reasons, there has been an influx of mathematically oriented workers over the past fifty years into a subject that had been primarily an empirical one. Those workers will maintain momentum of the field, even as the gradual development of alternative sources of energy and methods of propulsion erodes funding. Various specific areas in which relatively short-term advances may be anticipated have been indicated throughout the text. It is of interest here first to offer a summary list of topics that in our opinion should enjoy progress over a somewhat longer term, but less than a fifty-year period; a similar list from an individual perspective has been previously published [205].

Reduced chemistry and rate-ratio asymptotics will be extended to include premixed flames, diffusion flames, ignition and detonation, eventually encompassing higher hydrocarbons, propellants, pollutants, toxics, and flame inhibition. The methods of rate-ratio asymptotics also will be applied to problems of flame stability, flame propagation in nonuniform flows, and diffusion flame dynamics. Multiple-scale expansions will be developed for detonation transmission and failure. Acoustic interactions of laminar and turbulent gaseous and multiphase flames will be described well theoretically. Knowledge of the dynamics of subcritical and supercritical droplet and spray combustion will be improved. Clarifications will be obtained of coflow and counterflow flame spread along charring fuels, liquid fuels, and nonhomogeneous fuel beds. Specific conditions will be established under which lifted flames are dominated by premixing or by diffusion flamelets under both laminar and turbulent conditions. Physically well-justified subgrid-scale models will be developed for LES. Improved understanding of premixed turbulent combustion will be obtained through modeling with increased help from DNS. Fundamental advances will be made in describing the conditional scalar dissipation rate in nonpremixed combustion. All of these advances will continue to require close coordination with experiment.

In the longer term, a relevant query concerns whether new effective analytical methods will emerge to tackle the pertinent mathematical problems of combustion theory—methods comparable with the "asymptotic revolution" of the 1960s. Acknowledging haziness of fifty-year telescopes, we nevertheless, see no hint of such a development. Instead, we think that increasing interaction between theory and computation affords the best chance for very long-term advancement. There will surely be dramatic improvements in experimental capabilities, for example with laser diagnostics, and it will be important that theory keep contact with experiment for it to progress properly. The rate of development of computational capabilities is, however, greater, and since the underlying conservation equations are presumably known, advances through relationships with computation are most promising.

Numerical simulations are currently constrained by insufficient computational power, inability to provide the required spatial and temporal resolution in multidimensional systems with large disparities among the scales involved. The formidable difficulties in DNS of deflagration-to-detonation transition even in smooth-walled tubes are an example. Further development of computing facilities, however, assuredly will occur, and it is not unreasonable to predict revolutions in this area, whether through incremental advances in parallel computing, through quantum-mechanical computing, through DNA computing or through a strategy as yet unknown. It is not far-fetched to hope for increases in computing power by orders of magnitude over what is in place today, during the next fifty years, making possible three-dimensional, unsteady calculations with detailed chemistry, perhaps even to the extent that subgrid-scale modeling becomes unnecessary at turbulence Reynolds numbers of practical interest. When such capabilities become available,

appropriately designed numerical experiments can help one to identify new structural and dynamic patterns, and to serve as a guide in breaking up the overall picture into interacting elementary building blocks, reducing the complexity to a simplicity that is consistent with human understanding. If scientific computation in combustion is still deemed of value at that time, then future combustion theorists will have vast resources for extending the body of intuitive combustion knowledge.

One exciting hope, with enormous consequences, is that elementary reaction rates, whether purely gas-phase or heterogeneous, may then be calculable from first principles of quantum mechanics, perhaps with a higher accuracy than they can be measured. Coupled with corresponding capabilities of calculating thermodynamic, molecular transport and radiative properties, including radiation transport, the world in which combustion occurs would become entirely accessible computationally, from the intricacies of propellant combustion to the complexities of soot emissions. The challenge to the combustion theoretician would then be to make the calculable results comprehensible, thereby helping us to solve the mystery of combustion.

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